Highly efficient Hydrogen evolution reaction by strain and phase engineering in composites of Pt and MoS$_2$ nano-scrolls

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Figure S1. TEM images of the dispersed MoS$_2$ sheets.

The concentration of the final exfoliated MoS$_2$ was measured by using the vacuum filter through a weighed membrane. The multi-layer MoS$_2$ with size of typically several hundred nanometers is observed.
Figure S2. TEM images of MoS$_2$ scrolls with a low magnification.

The tube-like morphology scrolled by interaction of MoS$_2$ with LCAs. The TEM images at a low magnification represent the produced MoS$_2$ scrolls. TEM images of the synthesized MoS$_2$ scrolls with the continuously increasing vertical distance along an axis.
Figure S3. TEM images of MoS$_2$@Pt sheets produced by the attachment of Pt NPs to the MoS$_2$ surface.

TEM images of the synthesized MoS$_2$@Pt sheets with low magnification. Magnified TEM images of the few layer MoS$_2$ surface with homogeneous attachment of Pt nanoparticles with size of 3 nm. HR-TEM images of Pt NPs on MoS$_2$ surface with a corresponding lattice spacing.
Figure S4. TEM images of MoS$_2$ scrolls@Pt.
Figure S5. TEM images of MoS$_2$ scrolls@Pt produced by the attachment of Pt NPs to the MoS$_2$ scroll surface.

<table>
<thead>
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<th>Element</th>
<th>Wt%</th>
<th>Wt% Sigma</th>
<th>Atomic %</th>
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<tr>
<td>S</td>
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<tr>
<td>Mo</td>
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<td>2.86</td>
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<tr>
<td>Pt</td>
<td>10.43</td>
<td>0.84</td>
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<tr>
<td>Total:</td>
<td>100.00</td>
<td></td>
<td>100.00</td>
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Figure S6. Calculation of the strain energy for MoS$_2$ derivatives.

We now discuss that the observed behavior can be explained by considering the various MoS$_2$ derivatives induced uniaxial strain on Raman modes responsible for E$_{12g}$ and A$_{1g}$, correspondingly. The uniaxial strain-induced peak splitting for the E$_{12g}$ mode enables us to calculate parameters: the Grüneisen parameter, $\gamma$, and the shear deformation potential, $\beta$, and the solution of the secular equation for the E$_{2g}$ mode $\Delta\omega_{E_{2g},l}$

$$
\gamma_{E_{2g}} = -\frac{\Delta\omega_{E_{2g}}^+ + \Delta\omega_{E_{2g}}^-}{2\omega_{E_{2g}}^0 (1 - \nu)\varepsilon}
$$

(1)

$$
\beta_{E_{2g}} = \frac{\Delta\omega_{E_{2g}}^+ - \Delta\omega_{E_{2g}}^-}{\omega_{E_{2g}}^0 (1 + \nu)\varepsilon}
$$

(2)

$$
\Delta\omega_{E_{2g}}^\pm = -\omega_{E_{2g}}^0 \gamma_{E_{2g}} (1 - \nu)\varepsilon \pm \frac{1}{2} \beta_{E_{2g}} \omega_{E_{2g}}^0 (1 + \nu)\varepsilon
$$

(3)

Here, $\Delta\omega$ is the change of frequency in the Raman mode, $\omega_{E_{2g}}^0$ is the E$_{2g}$ peak position at zero strain, $\nu$ is Poisson’s ratio, and $\varepsilon$ is the induced uniaxial tensile strain. A Grüneisen parameter of 1.1 and a shear deformation potential of 0.78 for both the monolayer and bilayer MoS$_2$ attached to a substrate ($\nu = 0.33$) have been reported. In case of formation of MoS$_2$@Pt scrolls, these calculated parameters are used to estimate the strain applied to a free-standing MoS$_2$. By inserting $\omega_{E_{2g}}^0 = 380.9$ cm$^{-1}$ from the Raman spectra, $\gamma_{E_{2g}} = 1.1$, $\beta_{E_{2g}} = 0.78$, and a Poisson’s ratio of $\nu = 0.27$ for multilayer MoS$_2$ in Equation 3, we obtained the E$_{12g}$ peak shifts for with an ~4.9 cm$^{-1}$/% strain, and the E$_{12g}^+$ peak shifts for with an ~1.1 cm$^{-1}$/% strain. These results are nearly equal to the DFT calculated tensile strain on the
monolayer MoS$_2$ with the E$^{1\text{g}}$ peak shifts having a strain of $4.5 \text{ cm}^{-1} /\%$ and the straining monolayer and bilayer MoS$_2$ with strains of $4.5 \pm 0.3 \text{ cm}^{-1} /\%$ and $4.6 \pm 0.4 \text{ cm}^{-1} /\%$, respectively.$^2$
Figure S7. Raman spectra of the MoS$_2$ sheets located at 380.9 and 406.4 cm$^{-1}$ with FWHMs of 4.6 and 5.0 cm$^{-1}$, respectively.

The corresponding bands for MoS$_2$ sheets are located at 380.9 and 406.4 cm$^{-1}$ with FWHMs of 4.6 and 5.0 cm$^{-1}$, respectively. The $E_{12g}$ mode involves in-plane displacement and shear force of Mo and S atoms, whereas the $A_{1g}$ mode involves out-of-plane symmetric displacement and compressive force of S atoms along the c axis.
References


